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INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

<b>(51) International Patent Classification 6 :</b> C09J 123/08	<b>A1</b>	<b>(11) International Publication Number:</b> WO 97/15635 <b>(43) International Publication Date:</b> 1 May 1997 (01.05.97)
<b>(21) International Application Number:</b> PCT/EP96/03858 <b>(22) International Filing Date:</b> 2 September 1996 (02.09.96) <b>(30) Priority Data:</b> 9521499.5      20 October 1995 (20.10.95)      GB <b>(71) Applicant (for all designated States except US):</b> EXXON CHEMICAL PATENTS INC. [US/US]; 1900 East Linden Avenue, Linden, NJ 07036 (US). <b>(72) Inventors; and</b> <b>(75) Inventors/Applicants (for US only):</b> FAISSAT, Michel, Louis [FR/BE]; Avenue Marie-Louise 21, B-1410 Waterloo (BE). ROBBERECHTS, Herman, Diederik, Marie, Emiel [BE/BE]; Ipsvoordestraat 43, B-1880 Nieuwenrode (BE). <b>(74) Agents:</b> BAWDEN, Peter, Charles et al.; Exxon Chemical Limited, Exxon Chemical Technology Centre, Abingdon, P.O. Box 1, Oxfordshire OX13 6BB (GB).		<b>(81) Designated States:</b> CN, KR, US, European patent (AT, BE, CH, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).  <b>Published</b> <i>With international search report.</i>
<b>(54) Title:</b> SPRAYABLE ETHYLENE BASED HOT MELT ADHESIVES  <b>(57) Abstract</b>  This invention relates to sprayable hot melt adhesive compositions comprising a polymer of ethylene and a polar monomer and a tackifier.		

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## Sprayable Ethylene Based Hot Melt Adhesives

Inventors: Michel Faissat and Herman Robberechts

5 Field of the Invention

This invention relates to sprayable hot melt adhesives based upon ethylene copolymers and methods to apply these hot melt adhesives, particularly in disposable  
10 packaging and assembly applications and articles produced by such methods.

Background of the Invention.

15 Sprayable hot melt adhesives are desirable for the economic and commercial reasons that they can be thinly, evenly and quickly coated onto a substrate, saving in both time and material, and that they allow better substrate coverage per quantity of adhesive. Typical  
20 sprayable hot melt adhesives comprise an amorphous copolymer, generally a butene based copolymer of a styrene based block copolymer, combined with a tackifier. For example, U.S. Patent 4,959,207 discloses sprayable hot melt adhesives of butene copolymers having  
25 up to 5.5 to 10 weight % ethylene, tackifier and amorphous diluent having a softening point greater than 90 °C. Similarly, EP 442 045 A2 discloses sprayable hot melt adhesives of amorphous polyalphaolefins having 3 to 75 weight % C<sub>4</sub> to C<sub>10</sub> alpha-olefin, 25 to 95 weight %  
30 propylene 0 to 20 weight % ethylene. In the past attempts have been made to produce sprayable ethylene  $\alpha$ -olefin copolymer based hot melt adhesives. However, these blends were unsuccessful because the polymer would coagulate at the nozzle head or give uneven coating  
35 patterns of spots of unequal size rather than a regular even coating pattern. JP- 4180981 A addresses the problem of coagulation at the nozzle head by disclosing

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a hot melt adhesives of wax, bivalent phenol stabilizer, partially maleated or fumarated rosin ester tackifier and ethylene copolymer of ethylene mono-carboxylic vinyl ester/acrylic ester and suggesting that they will not  
5 clog the nozzle head when sprayed. However, when actually sprayed these hot melt adhesives will not produce a regular even pattern.

Similarly, WO 94 10256 discloses ethylene copolymer  
10 blends for hot melt adhesives. However these blends are not suggested for use in spraying applications.

Thus, this invention provides new sprayable hot melt adhesive compositions based upon ethylene copolymers.

15

#### Summary of the Invention.

This invention relates firstly to sprayable hot melt adhesive compositions comprising: a tackifier and an  
20 ethylene polymer having up to 45 weight % polar comonomer, based upon the weight of the copolymer and secondly to a process for spraying ethylene polymers comprising selecting a hot melt adhesive composition described above; provided that the tackifier should not  
25 be a rosin ester based tackifier with a softening point of 110-140 °C used in combination with a di-hydric phenol based stabilizer.

atomizing, spiral spraying or melt blowing the hot melt adhesive selected onto a substrate.

30

#### Detailed Description

In a preferred embodiment, this invention relates to sprayable hot melt adhesive compositions comprising: a  
35 tackifier, and an ethylene polymer having up to 40 weight % of a polar comonomer, based upon the weight of the copolymer; and melt index of 5 or more.

Preferred ethylene polymers that can be used in the practice of this invention include ethylene polymers having up to 40 weight %, from 0.5 to 33 weight %, even more preferably from 10 to 30 weight %, based upon the weight of the copolymer, of one or more polar comonomers. Preferred polar comonomers may be an ester, a carboxylic acid, an acrylic, and acrylic ester, a vinyl ester or the like. In a preferred embodiment the polar monomer is a vinyl ester or an alkyl acrylic ester, such as vinyl acetate, acrylic acid, methyl acrylate, ethyl acrylate, propyl acrylate, butyl acrylate, methacrylic acid, ethacrylic acid, and derivatives thereof.

In another preferred embodiment the ethylene polymers used in this invention have a melt index as measured by ASTM 1238, condition E, of 5 dg/min or more, preferably 15 dg/min or more, even more preferably 30 dg/min or more, even more preferably between about 100 and 10,000 dg/min.

In another preferred embodiment, the ethylene polymers of this invention are at least 50 % amorphous, preferably between 60 and 100% amorphous, even more preferably between 70 and 100% amorphous. Percent amorphous content is determined by Differential Scanning Calorimetry measurement according to ASTM E 794-85.

Preferred tackifiers that may be blended with the ethylene copolymers described above are those typically used in the art. Examples include, but are not limited to, aliphatic hydrocarbon resins, aromatic modified aliphatic hydrocarbon resins, hydrogenated polycyclopentadiene resins, polycyclopentadiene resins, gum rosins, gum rosin esters, wood rosins, wood rosin esters, tall oil rosins, tall oil rosin esters,

polyterpenes, aromatic modified polyterpenes, terpene phenolics, aromatic modified hydrogenated polycyclopentadiene resins, hydrogenated aliphatic resin, hydrogenated aliphatic aromatic resins, hydrogenated terpenes and modified terpenes, and hydrogenated rosin esters. In a preferred embodiment the tackifier is hydrogenated. Provided, however, that the tackifier should not be a rosin ester based tackifier with a softening point of 110-140 °C used in combination with a di-hydric phenol based stabilizer. Preferred tackifiers include the tall oil rosin esters and the cyclic C<sub>5</sub> based tackifiers.

The tackifier is typically present at about 15 weight % to about 75 weight %, based upon the weight of the blend, more preferably 30 weight % to 65 weight %, even more preferably 40 weight % to 60 weight %.

The ethylene polymer and the tackifier may be blended together by methods known in the art. For example the ethylene copolymer and the tackifiers can be physically admixed, they can be melt blended, they can be master batched, they can be solvent blended and the like. In one non-limiting example, the polymer and the tackifier are melted in a Z-blade mixer or in a Brabender twin screw extruder and blended under high shear conditions. Their blend components should be chosen so that a final blend viscosity of 20,000 mPaS or less is obtained, preferably 15,000 or less, even more preferably 10,000 or less, even more preferably 8,000 or less even more preferably between 500 and 8000. The range of 500 to 5000 mPaS at 180°C is ideal for spraying a low coating weight and wide spray pattern (spiral, net like web, or nebulized droplets).

In addition to the polymer and the tackifier, one can add another polymer. Preferred polymers include any of



the polymers described above as ethylene polymers. The second polymer may be present at up to 50 weight %, preferably 0.1 weight % to 30 weight %. A preferred class of polymer to be added to the composition is an ethylene/propylene/butene terpolymer. Ethylene/propylene/ butene terpolymers are not typically sprayable, however, when blended with the ethylene polymers described above, they can be formed into sprayable compositions. Examples of preferred ethylene/propylene/butene terpolymers are VESTOPLAST™ 708, VESTOPLAST™750, and VESTOPLAST™408 which are commercially available from Hüls. The ethylene/propylene/butene terpolymer is preferably present at up to 50 weight %, even more preferably at 0.1 weight % to 20 weight %, even more preferably at 0.1 weight % to 15 weight %.

Another optional component of the ethylene polymer/tackifier composition is a plasticizer. Preferred plasticizers include mineral oils, polybutenes, phthalates and the like. Particularly preferred plasticizers include phthalates such as diisoundecyl phthalate (DIUP), diisononylphthalate (DINP), dioctylphthalates (DOP) and the like. Another optional component of the ethylene polymer/tackifier composition is a low molecular weight product such as wax, oil or low Mn polymer, (low meaning below Mn of 5000, preferably below 4000, more preferably below 3000, even more preferably below 2500). Preferred oils include aliphatic napthenic oils. Preferred low Mn polymers include polymers of lower alpha olefins such as propylene, butene, pentene, hexene and the like. A particularly preferred polymer includes polybutene having an Mn of less than 1000. An example of such a polymer is available under the trade name PARAPOL™ 950 from Exxon Chemical Company. PARAPOL™ 950 is an liquid

polybutene polymer having an Mn of 950 and a kinematic viscosity of 220cSt at 100 °C, as measured by ASTM D 445.

The blend may then be applied directly to a substrate as a hot melt adhesive or may be sprayed thereon. Spraying is defined to include atomizing, spiral spray controlled fiberization and melt blown techniques. Melt blown techniques are defined to include the methods described in U.S. patent 5,145,689 or any process where air streams are used to break up filaments of the extrudate and then used to deposit the broken filaments on a substrate. In general, melt blown techniques are processes that use air to spin hot melt adhesive fibers and convey them onto a substrate for bonding. Fibers sizes can easily be controlled from 20-200 microns by changing the melt to air ratio. Few, preferably no, stray fibers are generated due to the inherent stability of adhesive melt blown applicators. Under UV light the bonding appears as a regular, smooth, stretched dots pattern. Atomization is a process that uses air to atomize hot melt adhesive into very small dots and convey them onto a substrate for bonding.

The blend may also be applied to a substrate using conventional techniques for applying a hot melt adhesive to a substrate. Examples include hot melt jet nozzle coating, hot melt multiline coating, hot melt slot die coating, hot melt wheel coating, hot melt roller coating, and solvent roller coating.

In a preferred embodiment the blend is applied to a substrate using a spiral spray. Spiral spray is a method used for producing a filamentary spiral application. In one example the hot melt adhesive and spray air are mixed inside the nozzle, eliminating external influences on the spray pattern. In another example, the hot melt adhesive is drawn into a small

fiber by high-velocity air jets. The fiber is then rotated by jets until it contacts the substrate, producing a helical pattern from the single strand of adhesive.

5

The sprayed hot melt adhesives of this invention can be used for disposable diaper and napkin chassis construction, elastic attachment in disposable goods converting, packaging, labelling and other assembly applications. Particularly preferred applications include: baby diaper leg elastic, diaper frontal tape, diaper standing leg cuff, diaper chassis construction, diaper core stabilization, diaper liquid transfer layer, diaper outer cover lamination, diaper elastic cuff lamination, feminine napkin core stabilization, feminine napping adhesive strip, industrial filtration bonding, industrial filter material lamination, filter mask lamination, surgical gown lamination, surgical drape lamination, and perishable products packaging.

20

#### Tests and Materials.

Melt Index (MI) was measured according to ASTM 1238, condition E (dg/min).

25 Density was measured according to ASTM D-792 (g/cc).

T-Peel was measured ASTM D1876-72 (20 mm wide test specimens and 2 inch/min (5.08 cm/min) head speed)

Viscosity was measured according to ASTM D 3236 at 180 °C (mPaS)

30 Penetration was measured according to ASTM D1321-86.

SAFT (Shear Adhesion Fail Temperature) was measured by adhering a coated polyethylene strip of 25 mm width to stainless steel via press lamination with a contact area of 12.5mm x 25mm, hanging samples in an oven held at 25 °

35 C and suspending a 500 gram weight from the bottom of the strip. The temperature is raised at 0.4 °C/min and

the bond failure temperature is measured. The SAFT is the average of three test samples. (°C).

Hot Shear was measured by suspending a 1000 gram weight from a 25 mm wide strip of polyethylene film coated with the adhesive formulation which is adhered with press lamination to a stainless steel plate with a contact area of 12.5mm x 25mm. The sample is placed in a ventilated oven at 40 °C. Every 30 minutes the temperature is increased by 10°C. Time and temperature are recorded until stress failure occurs. Typically these tests are made and recorded individually to determine the reliability of the holding power. (min, °C)

Sprayability was measured on a CT 325 Meltex coater under the conditions listed in Example 1. Perfect spiral is shown in figure 1, acceptable spiral is shown in figure 2 and no spiral but unequal spots is shown in figure 3.

Static shear was measured according to hot shear test set-up described above, but temperature is maintained constant at 50 °C. Time is recorded until stress failure occurs.

Molecular weights (Mw and Mn) were measured by Gel Permeation Chromatography, unless otherwise noted, using a Waters 150-CV Gel Permeation Chromatograph equipped with a differential refractive index (DRI) detector and polystyrene standards. Polymer samples were dissolved in trichlorobenzene solvent at 165 °C and the solutions were run at 145 °C using three Shodex GPC AT-80 M/S columns in series. This general technique is discussed in "Liquid Chromatography of Polymers and Related Materials III" J. Cazes Ed., Marcel Decker, 1981, page 207, which is incorporated by reference herein. No corrections for column spreading were employed; however, data on generally accepted standards, e.g. National Bureau of Standards Polyethylene 1475, demonstrated a precision with 0.1 units for Mw/Mn which was calculated

from elution times. The numerical analyses were performed using Expert Ease™ software available from Waters Corporation.

Melt Flow Rate (MFR) was measured according to ASTM D  
5 1238 (10 kg, 230 °C)

VECTOR™ 4461D is a styrene-butadiene-styrene block copolymer having an melt flow rate of 23 dg/min, a styrene content of 43 weight %, and an Mw of 60,000.

10 VECTOR™ 4113 is a styrene-isoprene-styrene block copolymer having an melt flow rate of 10 dg/min, a styrene content of 15 weight %, and an Mw of 143,000.  
XW-23 is an ethylene/butyl acrylate copolymer having an MI of 320 dg/min, and a butyl acrylate content of 33  
15 weight %.

PARAPOL™ 950 is a liquid polybutene having an Mn of about 950 and a Kinematic viscosity of 220 cst at 100 °C, as measured by ASTM D445.

20 ESCOREZ™ 5380 is a hydrogenated polycyclopentadiene tackifier having a softening point of 85 °C.

ESCOREZ™ 5320 is a hydrogenated polycyclopentadiene tackifier having a softening point of 125 °C.

ESCOREZ™ 5300 is a hydrogenated polycyclopentadiene tackifier having a softening point of 105 °C.

25 ESCOREZ™ 6368 (ECR 368LC) is a hydrogenated aliphatic aromatic tackifier having a softening point of 100 °C.

OPTEMA™ XS 9304 is an ethylene/methylacrylate copolymer having and MI of 270 dg/min and density of 0.939 g/cc and an MA content of 21.5 %.

30 OPTEMA™ TC 140 is an ethylene/methylacrylate copolymer having and MI of 135 dg/min and density of 0.939 g/cc and an MA content of 21.5 %.

ADEVA™ 629 is a high modulus ethylene/vinyl acetate copolymer having an MI of 400 dg/min, and a VA content  
35 of 33%.

ESCORENE™ UL 40028 is an ethylene/vinyl acetate copolymer having an MI of 400 dg/min, and a VA content of 28%.

5 ESCORENE™ UL 15028 is an ethylene/vinyl acetate copolymer having an MI of 150 dg/min, and a VA content of 28 %.

ESCORENE™ UL 05540 is an ethylene/vinyl acetate copolymer having an MI of 55 dg/min, and a VA content of 40%

10 ECR 404 is a light color hydrogenated polycyclopentadiene tackifier having a softening point of 118 °C.

VECTOR™ DPX 554 is a styrene-isoprene-styrene block copolymer having an MFR of 9 and a styrene content of 30.  
15 weight %.

VESTOPLAST™750 is an ethylene/propylene/butene copolymer available from Hüls, having a density of 0.87 and an MV of 70,000.

20 VESTOPLAST™708 is an ethylene/propylene/butene copolymer available from Hüls, having a density of 0.87 and an MV of 45,000.

DIUP is diisoundecyl phthalate

PRIMOL™ 352 is a hydrogenated white aliphatic naphthenic oil available from Exxon Chemical Company.

25 Lotryl HY™ 6190 is an ethylene butyl acrylate copolymer having an MI of 400 dg/min and a butyl acrylate content of 28% available from Atochem.

EX 659 is a high modulus ethylene/vinyl acetate copolymer having an MI of 800 dg/min, and a VA content  
30 of 28% available from Exxon Chemical Company.

Meltex CT 325 is a coating machine from Nordson/Meltex.

EP 34-6SD spray head is a spiral spray coating head.

IRGANOX™ 1010 is an antioxidant available from Ciba  
Geigy described in U.S. Patents 3,285,855 and 3,644,482.

35

Examples

Several different ethylene copolymers were blended with one or more tackifiers according to the following procedure. In a Z-blade mixer with the rotorblades in Z-shape, 204 grams of the ethylene copolymer and 276 grams of tackifying resin were blended incrementally at 180 °C, under nitrogen, until the total volume of the polymer and tackifier were mixed. The components were allowed to mix for 10 minutes, then the parapolymer DIUP, etc, if any, were added. The blend was then mixed for another 50 minutes and poured into a release paper container. Several tests were performed on the various blends according to the procedures described above and the blends were sprayed using a CT 325 Meltex coater under the following conditions:

Melt section temp.: 170°C

Hoze temp.: 175°C

Nozzle temp.: 175°C

Air temp.: 200°C

12 notches spiralhead

Nip roll: 1 bar

Brake roll 1: 0-0.5 bar

Brake roll 2: 0-0.5 bar

Unwind 1: 0-0.5 bar

Lamination roll 1: 4 bar

Lamination roll 2: 4 bar

Unwind 2: 0-0.5 bar

Cooling Unit: 15-16°C

Web speed: 125 m/min

pump speed between 40-50 rpm on TACHO to have a coating

weight of  $\pm 0.5$  g/lm for a spiral diameter of 4-4.5 cm

The polymers, blend components and data are reported in Tables 1 to 4.

Symbols and terms in the Tables

1 = g/spiral spray  $\pm 4$ cm, Adhesive spiral sprayed onto a nonwoven coverstock of thermally bonded polypropylene and then bonded to a polyethylene substrate.

2 = Adhesive coated onto polyethylene substrate then bonded to another polyethylene substrate.

3 = adhesive meltblown onto a nonwoven coverstock of thermally bonded polypropylene then bonded to polyethylene substrate.

Perfect blownability means that the adhesive was deposited evenly with the same coating weight left to right on the substrate.

Perfect spiral means that the spiral is a wide and regular spiral of about 4 cm.



TABLE 1

	Blend AA	Blend BB	Blend CC	Blend DD
Adeva 629	-	-	40	-
EX659	-	-	-	25
Escorene UL 40028	40	40	-	-
Escorene UL 05540	-	-	-	15
ECR-368LC	40	40	40	40
Parapol 950	20	15	15	15
DIUP	-	5	5	5
Irganox 1010	0.5	0.5	0.5	0.5
Sprayability 4- 4.5 cm spiral width	perfect spiral	perfect spiral	perfect spiral	perfect spiral
Sprayability 2- 2.5 cm spiral width	perfect spiral	perfect spiral	perfect spiral	perfect spiral
T-Peel NWC/PE @ R.T. <sup>1</sup>	610 g/spray	515 g/spray	620 g/spray	600 g/spray
Failure mode	adhesive	adhesive	cohesive	adhesive
T-Peel NWC/PE @ 40 °C <sup>1</sup>	80 g/spray	80 g/spray	50 g/spray	60 g/spray
Failure mode	adhesive	adhesive	adhesive	adhesive
blend viscosity @ 180 °C (mpas)	1420	1280	1360	1540

TABLE 2

	Blend EE	Blend FF	Blend GG
Vector 4461D			20
XW-23	40	40	-
DIUP	5	5	-
Escorene UL 40028			5
ECR-368LC	40		55
Escorez 5380		40	-
Parapol 950	15	15	-
Primol 352			20
Irganox 1010	.05	0.5	0.5
Sprayability 4-4.5 cm spiral width	perfect spiral	perfect spiral	perfect spiral
Sprayability 2-2.5 cm spiral width	perfect spiral	perfect spiral	perfect spiral
T-Peel NWC/PE @ R.T. <sup>1</sup>	180 g/spray	190 g/spray	295 g/spray
Failure mode	adhesive	adhesive	adhesive
T-Peel NWC/PE @ 40 °C <sup>1</sup>	10 g/spray	20 g/spray	130 g/spray
Failure mode	adhesive	adhesive	adhesive
blend viscosity @ 180 °C (mpas)	1550	1650	1325
Hot shear (min, °C)	29,40 30,40 25,40	2,50 3,50 30,40	
Static shear @ 50 °C(min)	4	4	
SAFT(°C)	49/50/48	47/48/48	
Penetration (mm) @ R.T.	5.18	5.82	
Penetration (mm) @ 40°C	10.7	12.4	
T-Peel PE/PE <sup>2</sup> (g/cm)@ R.T.	490 CF	335 CF	
T-Peel PE/PE <sup>2</sup> (g/cm)@ 40°C		25 CF	

TABLE 3

	Blend HH	Blend II	Blend JJ
Lotryl HY 6190	-	-	40
Optema XS 9304	-	40	-
Escorene UL 40028	5	-	-
Vector DPX 554	20	-	-
ECR-368LC	27.5	40	40
Escorez 5300	27.5	-	-
Parapol 950	-	15	15
Primol 352	20	-	-
DIUP	-	5	5
Irganox 1010	0.5	0.5	0.5
Sprayability 4-4.5 cm spiral width	Perfect spiral	Perfect spiral	Perfect spiral
Sprayability 2-2.5 cm spiral width	Perfect spiral	Perfect spiral	Perfect spiral
T-Peel NWC/PE @ R.T. <sup>1</sup>	400 g/spray	570 g/spray	285 g/spray
Failure mode	adhesive	adhesiv	cohesive
T-Peel NWC/PE @ 40 °C <sup>1</sup>	120 g/spray	60 g/spray	60 g/spray
Failure mode	adhesive	adhesive	adhesive
blend viscosity @ 180 °C (mpas)	1050	1825	1955

TABLE 4

	Blend KK	Blend LL	Blend MM	Blend NN
Vector 4113	25	-	-	-
Optema TC 140	-	-	-	40
XW-23	-	-	45	-
Escorene UL 15028	-	40	-	-
ECR 404	55	-	-	-
Escorez 5320	-	-	40	-
Escorez 6368 (ECR 368 LC)	-	40	-	40
Parapol 950	-	20	15	20
Primol 352	20	-	-	-
Irganox 1010	0.5	0.5	0.5	0.5
Sprayability 4-4.5cm spiral width	Perfect	Not Accept- able	Good	Good
Sprayability 2-2.5cm spiral width	Perfect	Good	Perfect	Perfect
T-Peel NWC/PE @ R.T. <sup>1</sup>	410 g/spray	860 g/spray	500 g/spray	585 g/spray
Failure mode	AF	AF	CF	AF
T-Peel NWC/PE @40°C <sup>1</sup>	200	170	120	90
Failure mode	AF	AF	CF	CF
blend viscosity @ 180 °C (mpas)	4500	2800	3600	3400
Hot shear (min, °C)	18,60 18,60 6,60	7,50 8,50 7,50	24,50 22,50 27,50	3,50 4,50 1,50
Static shear @ 50°C (min)	98	8	26	11
SAFT(°C)	81/80	51/50/49	59/59/60	49/52/53
Penetration(mm)@R.T.	9.85	2.87	2.45	2.55
Penetration(mm)@40°C	12.4	7.61	7.08	6.55
T-PEEL PE/PE <sup>2</sup> (g/cm)@ R.T.	890 CF	1215 CF	860 CF	1140 CF
T-PEEL PE/PE <sup>2</sup> (g/cm)@ 40°C	575 AF	425 CF	110 CF	265 CF
Blownability	perfect	perfect	perfect	perfect
T-PEEL NWC/PE <sup>3</sup> "Melt Blown" (N/25mm)@R.T.	1.75 AF	1.15 AF	1.15 AF	1.05 AF
T-PEEL NWC/PE <sup>3</sup> "Melt Blown" (N/25mm)@40°C	0.65 AF	0.2 AF	0.3 AF	0.2 AF

All documents described herein are incorporated by reference herein, including any priority documents and/or testing procedures. As is apparent from the foregoing general description and the specific  
5   embodiments, while forms of the invention have been illustrated and described, various modifications can be made without departing from the spirit and scope of the invention. Accordingly, it is not intended that the  
10   invention be limited thereby.

Claims:

1. A process for spraying ethylene polymers comprising selecting a hot melt adhesive composition comprising: a  
5 tackifier and an ethylene polymer having up to 45 weight % polar comonomer, based upon the weight of the copolymer and an MI of 5 or more; provided that the tackifier should not be a rosin ester based tackifier with a softening point of 110-140 °C used in combination  
10 with a di-hydric phenol based stabilizer.  
atomizing, spiral spraying or melt blowing the hot melt adhesive selected onto a substrate.
2. The process of claim 1 wherein the comonomer is an  
15 ester, a carboxylic acid, an acrylic, and acrylic ester, or a vinyl ester.
3. The process of any of the above claims wherein plasticizer is additionally present.  
20
4. The process of claim 6 wherein the plasticizer is a phthalate, preferably diisoundecyl phthalate.
5. The process of any of the above claims wherein an  
25 oil or wax is additionally present, preferably the oil is an olefinic or naphthenic oil.
6. The process of any of the above claims wherein a polybutene copolymer having an Mn of less than 2500 is  
30 additionally present.
7. The process of any of the above claims where the tackifier is aliphatic hydrocarbon resins, aromatic modified aliphatic hydrocarbon resins, hydrogenated  
35 polycyclopentadiene resins, polycyclopentadiene resins, gum rosins, gum rosin esters, wood rosins, wood rosin esters, tall oil rosins, tall oil rosin esters,

polyterpenes, aromatic modified polyterpenes, terpene phenolics, aromatic modified hydrogenated polycyclopentadiene resins, hydrogenated aliphatic resin, hydrogenated aliphatic aromatic resins,  
5 hydrogenated terpenes and modified terpenes, and  
hydrogenated rosin esters.

8. The process of any of the above claims wherein the  
tackifier is a hydrogenated, linear cyclic and/or  
10 branched pentene.

9. The process of any of the above claims further comprising a second copolymer.

## INTERNATIONAL SEARCH REPORT

International Application No  
PC1/EP 96/03858A. CLASSIFICATION F SUBJECT MATTER  
IPC 6 C09J123/08

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)  
IPC 6 C09J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US,A,5 310 803 (HANSEN DENNIS D) 10 May 1994 see column 1, line 16 - line 22 see column 6, line 22 - line 35 see column 10, line 58 - column 11, line 9 ---	1,2,7-9
X	WO,A,95 13332 (MINNESOTA MINING & MFG) 18 May 1995 see page 6, line 36 - page 9, line 19 see page 10, line 26 - line 30 ---	1-3,7-9
Y	---	5,6
X	WO,A,91 18043 (DU PONT) 28 November 1991 see page 4, line 25 - page 7, line 35 see page 9, line 4 - line 15 see page 11, line 9 - line 30 see page 12, line 9 - line 21 ---	1-5,7-9
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☒ Further documents are listed in the continuation of box C.☒ Patent family members are listed in annex.

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Date of the actual completion of the international search

16 December 1996

Date of mailing of the international search report

22.01.97

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PCT/EP 96/03858

## C. (Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	<p>WO, A, 92 12212 (EXXON CHEMICAL PATENTS INC)  23 July 1992  see page 9, line 27 - page 10, line 16  -----</p>	5, 6

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Information on patent family members

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